Predicting the Viscosity of Natural Gas¹

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ABSTRACT

Viscosity of the natural gas has been evaluated by four different methods;

namely Lohrentz-Bray-Clark (LBC) method, Pedersen et al. (PFCT) method, the

SUPERTRAP method and Vesovic-Wakeham (VW) method. The predictions have been

compared with the available experimental data that cover the temperature range from

240K to 444K and pressures up to 550 bars..

The PFCT and VW methods showed the smallest rms deviations, while the

predictions of SUPERTRAP were only marginally worse. The results indicate that these

three methods are capable of predicting the viscosity of natural gas with rms deviation

of 3% and maximum deviations of 5-6%. The LBC method proved less reliable with

maximum deviations of 8-9% observed.

1. INTRODUCTION

The increasing demand for natural gas has led to the need to develop a more reliable reservoir characterisation and simulation. The upstream gas industry, through the gas suppliers, is also being faced with increasing demand for precision in the monitoring of gas supplies. For the exploitation and usage to be optimal, an accurate and reliable knowledge of viscosity, along with other thermophysical properties of natural gas is a prerequisite. The diversity of possible natural gas mixtures and of conditions of interest precludes obtaining the relevant data by experimental means alone, thus requiring the development of prediction methods.

In principle, the viscosity of a fluid can be related to molecular motion and intermolecular forces acting among molecules. In practice, there is no rigorous theory that allows the complete evaluation of viscosity as a function of temperature and pressure in terms of realistic intermolecular potential. Thus any prediction method has to rely to a significant extent on a number of assumptions. Nevertheless, the need for greater accuracy and internal consistency of predicted viscosity data favours the prediction methods that are based on a theoretical framework and a stringent choice of primary data rather than those that are mere empirical correlations [1]. Modern developments in the field of prediction of viscosity of fluid mixtures have tended to follow two distinct routes [1]. Namely, methods developed that are based on the corresponding-states principle and methods developed that have a basis in kinetic theory. Embedded in each class of method is a mixing rule that allows the binary interaction to be estimated. It is at this level that most methods resort to assumptions that are difficult to verify *apriori*. In general modern methods do not require any

experimental information on mixture viscosity and can predict mixture viscosity from limited information on pure species properties. Their versatility, thermodynamic consistency and usually good accuracy makes them preferable to the empirical correlations of viscosity data that abound in the petroleum industry.

The existence of a number of different prediction methodologies [2-10] led us to perform a series of tests to find out what is the most suitable method for predicting the viscosity of the natural gas mixture. Although comparisons have been previously reported in the literature [11-12], recent publications of new measurements on the viscosity of natural gas and the developments in the predictive methodologies allows for a more comprehensive testing than has hitherto been possible. The choice of the prediction methods was limited to four, which encompass the methods employed in the petroleum industry and those that are at the forefront of recent developments in the field. Traditionally practitioners within the petroleum industry employ the Lohrentz-Bray-Clarke methodology [2] for estimating the viscosity of natural gas. This method is essentially empirical in nature and its present form is based on the experimental data available in the 1960's. Recently, methods that take advantage of the theoretical advances made in the thermophysical properties field have begun to make in-roads. The Pedersen et al. [3-4] method being the most popular choice, since it was specifically developed for hydrocarbon mixtures. Two further methods were chosen for the purposes of this work, one [5-6] based on the corresponding states principle and one based on the kinetic theory of fluid mixtures [7-9].

This paper reports on comparison of predictions of each method with the experimental viscosity data of natural gas mixture.

2. THEORY

In order to put the subsequent discussion of the results in a proper context it is useful to highlight and contrast the four methods chosen in this study. For brevity only essential elements of the four methodologies are presented here and the reader is referred, in each case to the original publications for more details.

2.1 Lohrenz-Bray-Clark (LBC) method

The LBC method [3] is still the most common method employed in the petroleum industry for estimating the viscosity of petroleum fluids. It is based on the observation that excess viscosity of a fluid is primarily a function of density and that to first approximation it can be assumed to be independent of temperature. The LBC method incorporates this finding into a general two parameter corresponding states methodology for estimating the viscosity of petroleum fluids from critical parameters of its constituent species. The viscosity of a fluid mixture is thus given by

$$\left[\left(\eta - \eta^0 \right) \right]^{\xi} + 10^{-4} \right]^{-0.25} = \sum_{j=0}^4 a_j \left(\frac{\rho}{\rho_c} \right)^j \tag{1}$$

$$\xi^{6} = \left[\sum x_{i} T_{ci}\right] \left[\sum x_{i} M_{i}\right]^{3} \left[\sum x_{i} P_{ci}\right]^{2}$$
 (2)

where x_i is the mole fraction of each pure species i, while T_{ci} , P_{ci} and M_i are the critical temperature, pressure and molecular weight respectively. The coefficients a_j are temperature independent and are those already suggested by Jossi *et al.* [13]. The zero density mixture viscosity is given by a simple mixing rule,

$$\eta^{0} = \frac{\sum_{i=1}^{N} x_{i} \eta_{i}^{0} M_{i}^{1/2}}{\sum_{i=1}^{N} x_{i} M_{i}^{1/2}}$$
(3)

while the critical density is calculated from the critical volume obtained from a mole fraction average mixing rule [3]. In principle the zero density viscosity, η_i^0 , of each pure species can be obtained from a number of reliable sources. In this work the original expressions employed by Lohrentz et al. [3] are used, where the viscosity is calculated by means of a simple corresponding states expression in terms of reduced temperature.

2.2 SUPERTRAP method

The method behind the SUPERTRAP computer programme [6] was originally developed by Ely and Hanley [5] and is based on the extended corresponding states principle. The viscosity of a given fluid mixture is calculated from the viscosity of the pre-defined reference fluid, by

$$\eta(T,\rho) = \eta_{ref.} (T/g,\rho h) \left[\frac{M}{M_{ref.}} \right]^{1/2} g^{1/2} h^{-2/3} X_{\eta}$$
 (4)

The quantity X_{η} is the correction factor for non-correspondence which for mixtures tends to be different from unity. The parameters g and h and the molecular weight of the mixture M are given by the following empirical mixing rules,

$$g = \frac{\sum_{i} \sum_{j} x_{i} x_{j} \sqrt{g_{i} g_{j}} h_{ij}}{h}$$

$$(5)$$

$$h = \sum_{i} \sum_{j} x_{i} x_{j} h_{ij} = \frac{1}{8} \sum_{i} \sum_{j} x_{i} x_{j} \left(h_{i}^{1/3} + h_{j}^{1/3} \right)^{3}$$
 (6)

$$M = \frac{\left[\sum_{i}\sum_{j} x_{i} x_{j} g_{ij}^{1/2} h_{ij}^{4/3} M_{ij}^{1/2}\right]^{2}}{g h^{8/3}}$$
(7)

For each pure species the parameters g_i and h_i can be calculated from the expressions that involve reduced temperature, reduced density and Pitzer acentric factors [5].

In the original implementation [5] the reference fluid was methane, but in the recent versions of the SUPERTRAP programme propane was chosen as the more appropriate representative. For the purposes of this work SUPERTRAP version 3.0 was used for all the calculations of the viscosity of natural gas.

2.3 PFCT method

Pedersen, Fredenslund, Christensen and Thomassen (PFCT) [3,4] modified the method of Ely and Hanley applying it specifically to the petroleum fluids. This method is nowadays gaining acceptance within petroleum industry as the method of choice for predicting the viscosity. The viscosity of the fluid mixture of interest is still calculated from the viscosity of the pre-defined reference fluid, but the reduced pressure rather than density is used as one of the corresponding states parameters. This approach avoids the need for estimation of mixture density from the equation of state, but the choice of pressure, rather than density, leads to difficulties of tackling the discontinuity in the viscosity at the saturation line.

In the PFCT method [3,4] the viscosity is calculated from the following equation

$$\eta(T,P) = \eta_{ref.} \left(\frac{T T_{cref.} \alpha_{ref.}}{T_{cmix.} \alpha_{mix.}}, \frac{P P_{cref.} \alpha_{ref.}}{P_{cmix.} \alpha_{mix.}} \right) \left[\frac{T_{cmix.}}{T_{cref.}} \right]^{-1/6} \left[\frac{P_{cmix.}}{P_{cref.}} \right]^{2/3} \left[\frac{M_{mix.}}{M_{ref.}} \right]^{2/3} \left[\frac{\alpha_{mix.}}{\alpha_{ref.}} \right]$$
(8)

where the parameters $T_{\rm cmix}$ and $P_{\rm cmix}$ are obtained from empirical mixing rules [3,4] that involve the critical temperature and pressure of each species in the mixture. The parameters α and $M_{\rm mix}$ account for the molecular size of different species. Both parameters have been treated as adjustable in the present method [3,4] and are given in terms of empirical relationships. Originally, methane was used as the reference fluid [3,4], but a more advanced version of the PFCT method exists [14] that is based on the two reference fluids (methane and decane). For natural gas, which is primarily methane, both versions are expected to give similar predictions and therefore a one-reference fluid version of the method was employed in this work.

2.4. Vesovic-Wakeham (VW) method

The VW methodology is based on rigid-sphere theory which is adequately modified to take into account the behaviour of real fluids in a self-consistent manner. The viscosity of a fluid mixture, η , is written in the form [7-9]

$$\eta = - \begin{vmatrix} H_{11} & \dots & H_{1N} & Y_1 \\ \vdots & & \vdots & \vdots \\ H_{N1} & \dots & H_{NN} & Y_N \\ Y_1 & \dots & Y_N & 0 \end{vmatrix} / \begin{vmatrix} H_{11} & \dots & H_{1N} \\ \vdots & & \vdots \\ H_{N1} & \dots & H_{NN} \end{vmatrix} + \kappa_{mix}$$
(9)

$$Y_i = x_i \left[1 + \sum_{j=1}^{N} \frac{m_j}{m_i + m_j} x_j \alpha_{ij} \overline{\chi}_{ij} \rho \right]$$
 (10)

$$H_{ii} = \frac{x_i^2 \overline{\chi}_{ii}}{\eta_i^0} + \sum_{j \neq i} \frac{x_i x_j \overline{\chi}_{ij}}{2 A_{ii}^* \eta_{ij}^0} \frac{m_i m_j}{(m_i + m_j)^2} \left[\frac{20}{3} + \frac{4m_j}{m_i} A_{ij}^* \right]$$
(11)

$$H_{ij} = -\frac{x_i x_j \overline{\chi}_{ij}}{2A_{ij}^* \eta_{ij}^0} \frac{m_i m_j}{(m_i + m_j)^2} \left[\frac{20}{3} - 4A_{ij}^* \right]$$
 (12)

$$\kappa_{mix} = \frac{15}{5\pi} \rho^2 \sum_{i=1}^{N} \sum_{j=1}^{N} x_i x_j \overline{\chi}_{ij} \alpha_{ij}^2 \eta_{ij}^0$$
 (13)

where η_{ij}^0 is the zero-density interaction viscosity, while A_{ij}^* is a weakly temperature dependent function for i-j pair interaction. The parameter α_{ij} accounts for the mean free path shortening for an i-j collision in the dense fluid, whereas $\overline{\chi}_{ij}$ is the pseudo-radial distribution function for the species i and j in the presence of all other species in the mixture. The pseudo-radial distribution function, $\overline{\chi}_{ij}$, is constructed by means of the following mixing rule which has no adjustable parameters,

$$\overline{\chi}_{ij}(\rho, T) = 1 + \frac{2}{5} \sum_{k=1}^{N} x_k (\overline{\chi}_k - 1) + \frac{6(\overline{\chi}_i - 1)^{1/3} (\overline{\chi}_j - 1)^{1/3}}{5[(\overline{\chi}_i - 1)^{1/3} + (\overline{\chi}_j - 1)^{1/3}]} \sum_{k=1}^{N} x_k (\overline{\chi}_k - 1)^{2/3}$$
(14)

while α_{ij} is obtained, for each isotherm, from

$$\alpha_{ij}(T) = \frac{1}{8} \left(\alpha_{ii}^{1/3} + \alpha_{jj}^{1/3} \right)^3 \tag{15}$$

The pseudo-radial distribution function for a pure component i is calculated from the viscosity of the pure species. In order to construct a realistic pseudo-radial distribution function that monotonically increases with increasing density, one is constrained in the choice of parameter α_{ii} . In fact, at each temperature, the requirement that the pseudo-radial distribution function is a continuous function of the molar density uniquely determines the value of parameter α_{ii} . This, in turn, allows for a unique determination of the pseudo-radial distribution function $\overline{\chi}_i$, as a function of the molar density at each isotherm for each pure component [7-9].

3. RESULTS

In order to make a sensible comparison of the predictions of the four methods it is important that the experimental data used are obtained in well-characterised apparatus with well-defined uncertainty limits. A literature search produced only three sets of [15-17] data that fulfil the above characteristics, highlighting the general issue of the scarcity of reliable measurements of the viscosity of fluid mixtures. Although a plethora of viscosity measurements must have been made in the routine industrial analysis, such data are usually not available in open literature and are not, in general, suitable for the comparison purposes due to lack of adequate characterisation.

It is customary to measure and report viscosity as a function of pressure at a given temperature. Three of the prediction methods used in this study, work on the basis of predicting the viscosity at a given temperature and density. Thus, it is necessary to convert temperature, pressure pairs into the appropriate temperature, density pairs. The density of the natural gas mixture was calculated by means of AGA8-DX92 correlation of Jeschke and Schley [18], which is based on an extended virial-type equation. The uncertainty of this correlation is ± 0.1 % for the temperature range from 265 to 335 K and pressures up to 12 MPa and ± 0.3 % for the other temperature and pressure ranges.

In the sixties Lee and his collaborators [15] performed extensive measurements of viscosity of natural gas. Four natural gas mixtures were examined and the measurements of viscosity were reported for temperatures from 311K to 444K and pressures from 13 bars to 550 bars. The measurements were performed by means of the capillary viscometer with an estimated accuracy of $\pm 2.0\%$ or better. The authors have

also performed the density measurements and in this case their quoted values are used as inputs in the three prediction methods that require temperature, density pairs.

Table 1 summarises the rms deviations obtained by comparing the predictions of the four methods with 111 available experimental data points. Overall agreement is very good. The PFCT, SUPERTRAPP and VW methods predict the experimental data equally well with rms deviation of approximately 2.8%. The LBC method produces a larger rms deviation, and closer examination of deviation plots indicates that for a number of isotherms the predicted density dependence of viscosity is not correct.

It is illustrative to examine in more detail the predictions of the four methods for each mixture. Mixture 1 is a nine component mixture, with the major components being methane ($x_{CH4}=0.863$), ethane ($x_{C2H6}=0.068$), carbon dioxide($x_{CO2}=0.068$), and propane ($x_{C3H8}=0.068$). As illustrated in Table 1, PFCT, SUPERTRAP and VW method predict the viscosity of this mixture equally well with the maximum deviations of 2.0%, 3.2% and -3.2% respectively. The LBC method in general underpredicts the viscosity with the maximum deviation of -5.7%. The closer examination of the deviation plots indicates the only systematic trends are observed at very low densities, up to 3000 moles.m⁻³, where all the methods predict a faster increase of viscosity with density than implied by the experimental data.

Mixture 2 is an eleven component mixture, with the major components being methane (x_{CH4} =0.717), ethane (x_{C2H6} =0.14), and propane (x_{C3H8} =0.83). Figure 1 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. The VW predict these data with the same rms deviation as for mixture 1, while the PFCT and SUPERTRAP tend to overpredict the data which results in an increase of rms deviations to 3.7% and 4.8% respectively. The largest deviations,

for PFCT and SUPERTRAP, of the order of 6-10%, are observed at the highest densities. The LBC method predicts a faster increase in viscosity, as a function of density, than is observed experimentally.

Mixture 3 is also an eleven component mixture, with the major components being methane ($x_{CH4}=0.807$), ethane ($x_{C2H6}=0.087$), nitrogen ($x_{N2}=0.048$) and propane ($x_{C3H8}=0.029$). The PFCT and SUPERTRAP predict these data with the similar rms deviations as for mixture 1, while the VW tends to underpredict the data which results in an increase of rms deviations to 3.1%. The maximum deviations observed for the three methods are 3.4%, 3.4% and -5.2% respectively. No systematic trends with density nor with temperature are observed and for brevity no deviation plot is included.

Mixture 4 is a ten component mixture, with the major components being methane (x_{CH4} =0.915), and ethane (x_{C2H6} =0.031). Figure 2 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. With such a large mole fraction of methane one would expect that the four prediction methods would have no problem predicting the viscosity of this mixture. In fact nearly the opposite is true and the rms deviations observed are larger than average, as illustrated in Table 1. Surprisingly, the maximum deviations are observed at low densities and for 411K isotherm are exceptionally large, ranging from 5-6% for SUPERTRAP to 10-12% for LBC method. This might indicate that for this particular mixture the uncertainty in the experimental viscosity might be higher than anticipated.

Recently Nabizadeh and Mayinger measured the viscosity of natural gas as a part of a more general programme of measuring the viscosity of hythane [16]. The measurements were carried out in an oscillating-disk viscometer with a claimed accuracy of $\pm 1\%$. Only one synthetic natural gas mixture (x_{CH4} =0.9467; x_{C2H6} =0.035;

 $x_{\rm N2}$ =0.0183) was examined and the viscosity measurements were reported in the temperature range 298K to 400K at pressures from atmospheric up to 71 bars.

Figure 3 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. The overall agreement is excellent. The LBC method tends to slightly underpredict the data, while the other three methods tend to overpredict the data. The rms deviations of LBC, PFCT, SUPERTRAPP and VW methods are 1.4%, 1.2%, 2.6% and 0.7% respectively, while the maximum deviations are –2.5%, 1.8%, 4.2% and 1.5% respectively. Thus, all the observed deviations are well within the combined accuracy of the experimental data and the accuracy of the methods. No systematic trends with temperature were observed and all four methods seem to predict the correct density dependence.

Assael and co-workers [17] also reported measurements of viscosity of natural gas. The measurements were carried out in a vibrating-wire viscometer with the claimed accuracy of $\pm 1\%$. Only one synthetic natural gas mixture (x_{CH4} =0.8484; x_{C2H6} =0.084; x_{C3H8} =0.005; x_{CO2} =0.0066; x_{N2} =0.056) was examined and the viscosity measurements were reported in the temperature range 240K to 353K at pressures from atmospheric up to 150 bars.

Figure 4 illustrates the deviations of the viscosity predicted by the four methods from the experimental data. The overall agreement is very good and the trends are similar to those displayed in Figure 3. The rms deviations of LBC, PFCT, SUPERTRAPP and VW methods are 0.9%, 1.9%, 3.4% and 1.5% respectively, while the maximum deviations are –2.4%, 3.1%, 5.9% and –3.8% respectively. No systematic trends with temperature were observed and all four methods seem to predict the correct density dependence.

Finally, a comparison was made with the viscosity data pertaining to a real reservoir fluid, as reported by Ali in Table 5 of his work [11]. The viscosity measurements are reported at one temperature, T=306K, and at pressures from 13.5 bars to 135 bars. No details of the experimental set-up and accuracy are available and the only justification for the inclusion of this data set is to extend the comparison to real reservoir fluids. In fact the presence of heavier components (x_{C6+} =0.005) in this mixture causes some retrograde condensation on lowering the pressure, thus making this fluid strictly speaking a condensate rather than natural gas. Nevertheless, the amount of liquid formed during the retrograde condensation is sufficiently small that it hardly affects the composition and the viscosity of the vapour phase.

All four prediction methods reproduce the nine experimental points very well. The rms deviations of LBC, PFCT, SUPERTRAPP and VW methods are 2.0%, 2.4%, 2.9% and 1.4% respectively, while the maximum deviations are –2.9%, 2.4%, 2.9% and –2.6% respectively. No systematic trends are observed and for brevity no deviation plot is included.

4. CONCLUSIONS

The four methods for predicting the viscosity of fluid mixtures, namely Lohrentz-Bray-Clark (LBC) method, Pedersen *et al.* (PFCT) method, the SUPERTRAP method and Vesovic-Wakeham (VW) method have been examined and used to predict the viscosity of natural gas. The predictions have been tested against the available experimental data. The comprehensive data of Lee *et al.* [15] containing 111 data points was reproduced by PFCT, SUPERTRAP and VW methods with rms deviation of

approximately 2.8%. This is only marginally worse than the claimed accuracy of the data, and adequate for most industrial requirements. The more accurate, but limited, data of Nabizadeh and Mayinger [16] and Assael et al. [17] was reproduced by PFCT and VW methods with rms deviation of less than 2%, while the SUPERTRAP exhibited larger rms deviation of the order of 3%. The LBC method performed relatively poorly, compared with the other three, reproducing the Lee *et al.* [15] data with rms deviation of 4.9%, while it did surprisingly well reproducing the more accurate data [16-17] only marginally worse than the claimed experimental uncertainty.

The viscosity of real reservoir condensate [11] was reproduced equally well by all the methods. The smallest deviations were exhibited by the VW method, while the largest were produced by SUPERTRAP.

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Table I The rms deviations of the experimental data of Ref. 15

	LBC	PFCT	SUPERTRAP	VW
	(%)	(%)	(%)	(%)
Overall	4.9	2.6	3.0	2.8
Mixture 1	4.2	1.0	1.4	1.9
Mixture 2	4.8	3.7	4.8	1.9
Mixture 3	5.4	1.9	1.7	3.1
Mixture 4	5.5	3.0	2.8	4.0

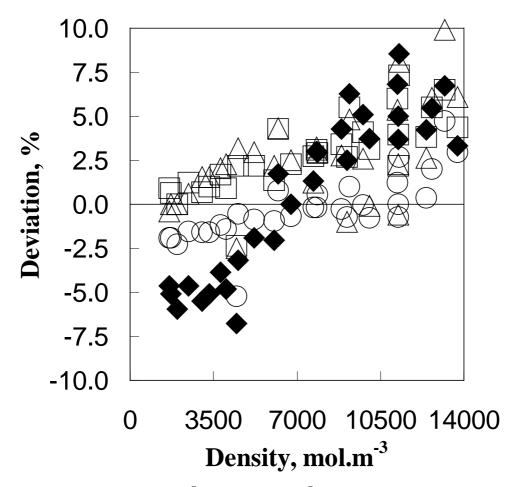


Figure 1. Deviations, $\left[\left(\eta_{p}-\eta_{exp.}\right)/\eta_{exp.}\right]x100\%$, of the predicted values, η_{p} , of the viscosity of natural gas from the experimental values, $\eta_{exp.}$, of mixture 2 [15] as a function of molar density, ρ .

[(\spadesuit) LBC method; () PFCT; (Δ) SUPERTRAP; (O) VW;]

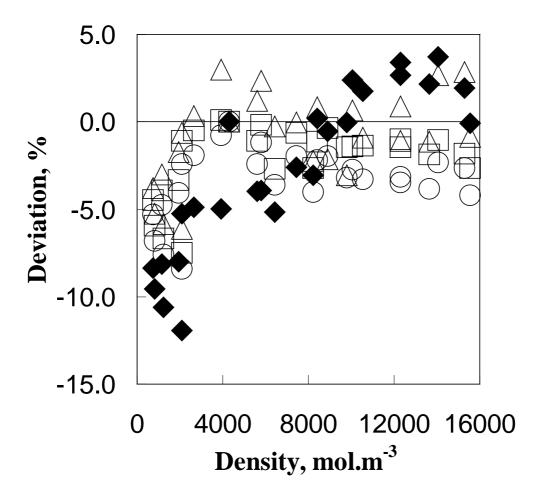


Figure 2. Deviations, $\left[\left(\eta_{p}-\eta_{exp.}\right)/\eta_{exp.}\right]x100\%$, of the predicted values, η_{p} , of the viscosity of natural gas from the experimental values, $\eta_{exp.}$, of mixture 4 [15] as a function of molar density, ρ .

[(\spadesuit) LBC method; () PFCT; (Δ) SUPERTRAP; (O) VW;]

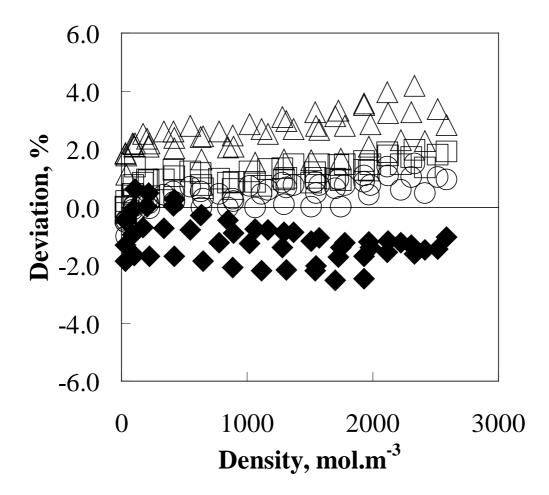


Figure 3. Deviations, $\left[\left(\eta_{p}-\eta_{exp.}\right)/\eta_{exp.}\right]x100\%$, of the predicted values, η_{p} , of the viscosity of natural gas from the experimental values, $\eta_{exp.}$, [16] as a function of molar density, ρ . [(\spadesuit) LBC method; () PFCT; (Δ) SUPERTRAP; (O) VW;]

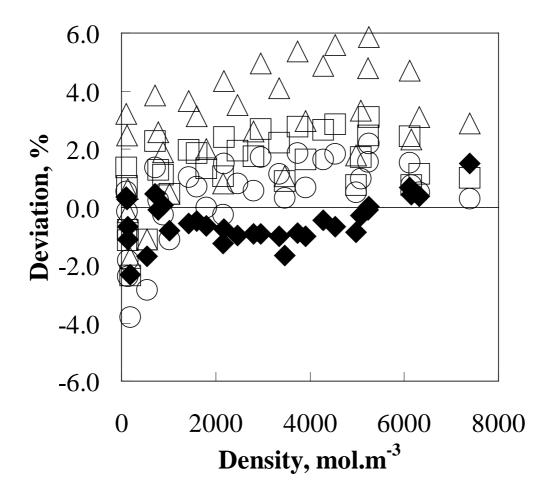


Figure 4. Deviations, $\left[\left(\eta_{p}-\eta_{exp.}\right)/\eta_{exp.}\right]x100\%$, of the predicted values, η_{p} , of the viscosity of natural gas from the experimental values, $\eta_{exp.}$, [17] as a function of molar density, ρ . [(\spadesuit) LBC method; () PFCT; (Δ) SUPERTRAP; (O) VW;]